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A REVIEW**

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
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DISTRIBUTION OF TRANSURANIC NUCLIDES IN SOILS: A REVIEW

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ABSTRACT

The literature is reviewed to ascertain the degree of movement and the distribution patterns for transuranic and uranium nuclides in soils. Typical plutonium and uranium profiles are presented and an attempt is made to identify unique characteristics causing deviation from an ideal distribution pattern. By far most of the distribution observations are with plutonium and little is reported for uranium and other transuranic nuclides.

INTRODUCTION

In the past decade interest has been growing in the environmental impact of certain transuranic radioisotopes because of their potentially hazardous nature to man. The potential for dispersion of these transuranics due to accidental release from components of the nuclear fuel cycle, improper radioactive waste management, accidental or purposeful nuclear weapon incidents, and further redistribution of existing global fallout and specific loci of contamination is being studied. There has been specific emphasis, during the past few years, on determining the degree of vertical migration of certain transuranic radioisotopes in soils, both in the laboratory and under field conditions. Further investigation on the environmental fate of transuranic elements is urgently needed in order to assess their long term distribution patterns.

This paper reviews past documentation on the vertical distribution of transuranic nuclides and uranium in the terrestrial environment under natural or near natural conditions. Observations made thus far will aid in predicting future redistribution rates and indicating where additional evaluations are needed.

REVIEWS

Francis (1973) and Price (1973) reviewed the literature through 1971 on plutonium and other transuranics in soils, plants, and animals. They found several works on plutonium and americium migration or distribution in soils, but information on other transuranics was lacking. Several papers described laboratory leaching studies on the movement of radioactive transuranic waste materials through soils. In general, these early papers showed that plutonium and americium can move through soils and are influenced by such factors as nature and oxidation state of source material, soil pH and organic matter content, presence of complexing agents, and cultural practices. It was suspected that, under natural conditions, most of the plutonium moved as colloidal or discrete dense plutonium oxide particles and not as solubilized species, the soluble plutonium being tightly bound by exchange mechanisms in the soil.

Wilhelmi (1974) reviewed the literature through 1973 and listed all the known sources of plutonium and transuranics added to the environment. Plutonium movement through the soil was discussed, and typical profiles, where available, were presented for fallout plutonium and for accidental plutonium releases at Rocky Flats, Colorado. Fallout plutonium was reportedly found to depths exceeding 30 cm, although the proportion of total profile plutonium found below 30 cm was only several percent. Wilhelmi (1974) pointed out that plutonium was strongly bound to soil material and migrated slowly; however, plutonium was found to migrate rapidly through cracks and fissures in geologic materials underlying soils. The review also revealed that there was little information on the influence of such factors as physical and chemical nature of soil and soil organic matter.

SOURCES OF TRANSURANICS

The source of transuranic nuclides in the environment, for purposes of discussion, can be categorized into global fallout, local fallout, safety shots, and accidental releases. Wilhelmi (1974) summarized the sources and noted that except for atmospheric nuclear explosions and the atmospheric burnup of a SNAP-9 power generator, almost all of the other sources were localized.

Global fallout is produced primarily from nuclear weapons explosions in the atmosphere, where substantial quantities of the radioactive debris are injected into the stratosphere. This material is slowly added to the environment over long periods of time and appears somewhat as a continuous source.

In contrast to global fallout is the local fallout associated with nuclear explosions and low level chronic releases from nuclear fuel processing. The local fallout from nuclear explosions and stack effluents from processing are deposited onto the soil surface soon after release and enter into the soil weathering processes immediately.

A number of nuclear weapons tests were conducted at the Nevada Test Site (NTS) and Tonopah Test Range (TTR) which resulted in little or no fission yield. These tests were chemical high explosive detonations of, or associated with, components of nuclear devices to determine the degree of safety or stability of the devices to the impact of an explosion. These tests were called "Safety Shots." The conduct of these tests resulted in the limited dispersion of the nuclear fuel, mostly plutonium and some americium as an impurity but in some cases natural or enriched uranium. The nuclear material was dispersed in the form of discrete metallic or oxide particles formed from the burning of the metal in air. The high temperatures attained in a nuclear explosion were not present; thus, the dispersed material had different characteristics from those it would have had if it had been involved in a nuclear explosion.

There have been a number of accidental releases of transuranic nuclides. Typical of accidental releases are the occasional low level releases from waste disposal facilities; accidental destruction of nuclear weapons as that which occurred over Palomares, Spain; and possible localized leaks of stored transuranic contaminated materials. Wilhelmi (1974) summarized all known accidental releases and noted that there were very few observations of transuranic nuclide distributions in soils associated with the accidental releases.

Expanding efforts to provide energy from nuclear sources will increase the inventory of transuranics and may increase the potential for an additional burden of transuranics in the environment. Those transuranic radionuclides of greatest concern are shown in Table 1 and Table 2, as compiled by Perkins (1975) and Langham (1971), respectively. Not all transuranic nuclides are considered hazardous, either because of the small amounts produced or the short radioactive half-lives.

A projected 100-fold increase in total curies of transuranics in waste over the next 24 years indicates the urgency for more information. However, this does not mean that there will be a 100-fold increase in the environmental burden of these transuranics.

DISTRIBUTION OF TRANSURANICS IN SOILS

There have been a number of studies dealing with the distribution of plutonium in various environmental situations, and only a few laboratory

Table 1. Activity of Heavy Elements in Accumulated Waste for Entire Nuclear Industry

Isotope	Normalized to 1 for ^{239}Pu						Decay Half-Life (y)
	1972	1976	1980	1986	1990	2000	
^{237}Np	0.22	0.22	0.20	0.15	0.14	0.76	2.1×10^6
^{239}Np	11.5	11	14.5	43.1	31.7	7.84	6.4×10^{-3}
^{236}Pu	0.00092	0.00075	0.00056	0.0036	0.010	0.012	2.85
^{238}Pu	36.1	49	55.6	167	283	361	8.64×10
^{239}Pu	1	1	1	1	1	1	2.44×10^4
^{240}Pu	1.54	1.7	1.89	4.45	5.33	3.14	6.58×10^3
^{241}Pu	315	300	295	403.3	367	255	1.32×10
^{242}Pu	0.0043	0.0044	0.0047	0.0099	0.011	0.011	3.79×10^5
^{243}Pu	--	--	--	--	--	--	--
^{241}Am	96.1	96.5	103	178	141	77.2	4.58×10^2
^{242m}Am	5.69	5.5	7.23	19.5	14.2	5.68	1.52×10^2
^{242}Am	5.69	5.5	7.23	19.5	14.2	5.68	1.82×10^{-3}
^{243}Am	11.5	11	14.5	43.1	31.7	7.84	7.95×10^3
^{242}Cm	5637	3020	2624	3183	953	278	4.45×10^{-1}
^{243}Cm	2.31	2.25	2.34	4.03	3.00	2.16	3.2×10
^{244}Cm	1561	1510	2441	9813	6464	919	1.76×10
^{245}Cm	0.23	0.23	0.50	2.64	1.92	0.33	9.3×10^3
^{246}Cm	0.046	0.046	0.072	0.49	0.35	0.063	5.5×10^3
^{247}Cm	--	--	--	--	--	--	--
TOTAL CURIES	7.2×10^5	1.4×10^7	7.9×10^7	9.3×10^8	1.2×10^9	1.8×10^9	

Adapted from Perkins (1975).

Table 2. Plutonium Economy of the Future

	Annual Production and/or in Use (kg)		
	1970-1980	1990-1990	1990-2000
^{239}Pu Power Production	20,000	60,000	80,000
^{238}Pu Space Applications	10-20	100	--
Medical Applications		5	6,000
Transplutonium Isotopes			
^{244}Cm	40	180	200
^{252}Cf	0.1	0.8	3.5

Adapted from Langham (1971)

studies designed to predict migration rates of plutonium and americium in specific soils. Of the several sources listed in the previous section, global fallout offers the most uniform source of plutonium and probably will offer the best source for long term migration studies.

Global Fallout

deBortoli and Gaglione (1969) sampled sandy or loamy sand soils from the environs of the Euratom Ispra Establishment, Ispra, Italy, in 1966 to determine the depth of fallout ^{239}Pu ($^{239},^{240}\text{Pu}$) penetration. The ^{239}Pu distribution in two profiles collected some distance apart are shown in Fig. 1, trace A. The ^{239}Pu was found to a depth of 15-20 cm in samples collected with a coring tool.

In 1970, Hardy and Krey (1971) measured ^{239}Pu ($^{239},^{240}\text{Pu}$) to a depth of 25-30 cm in a sandy soil in Brookhaven, NY (Fig. 1, trace B). This profile was also collected with a coring tool and compares closely with the profiles taken in 1966 from Ispra. In 1972, Hardy (1974) measured $^{239},^{240}\text{Pu}$ in a soil profile from North Eastham, MA., the first of a proposed time series of profiles (Fig. 1, trace C). This soil was not sampled by a coring method; rather, samples were obtained using a modification of the trench method used by the Nevada Applied Ecology Group (NAEG) at NTS (Fowler et al, 1974).

The use of coring tools for the purpose of collecting soil profile samples introduces a potentially serious cross-contamination variable. It is possible that both the similarity of profiles A and B, Fig. 1 and the amount of plutonium reported at depth, resulted from cross-contamination introduced by the coring tool. Profile C may reflect a smaller degree of cross-contamination; however, it has not been shown that the trench method prevents cross-contamination.

Hardy et al. (1972) reported ^{239}Pu ($^{239},^{240}\text{Pu}$) in soil profiles from widely separated locations within the United States. Less than 5% of the total ^{239}Pu was found in soil profile fractions below 30 cm. Some of these same profiles were reported by Bennett (1976).

McLendon (1975) reported the results of studies to determine the vertical distribution of ^{238}Pu and ^{239}Pu ($^{239},^{240}\text{Pu}$) in the on-plant and off-plant environs of the Savannah River Plant, Aiken, SC. In 1973, soil profile samples were collected by coring and slicing the cored soils into 0- to 5-, 5- to 15-, 15- to 22.5-, and 22.5- to 30-cm increments. Results indicated that in most cases the ^{238}Pu and ^{239}Pu were localized in the top two increments and remained relatively immobile.

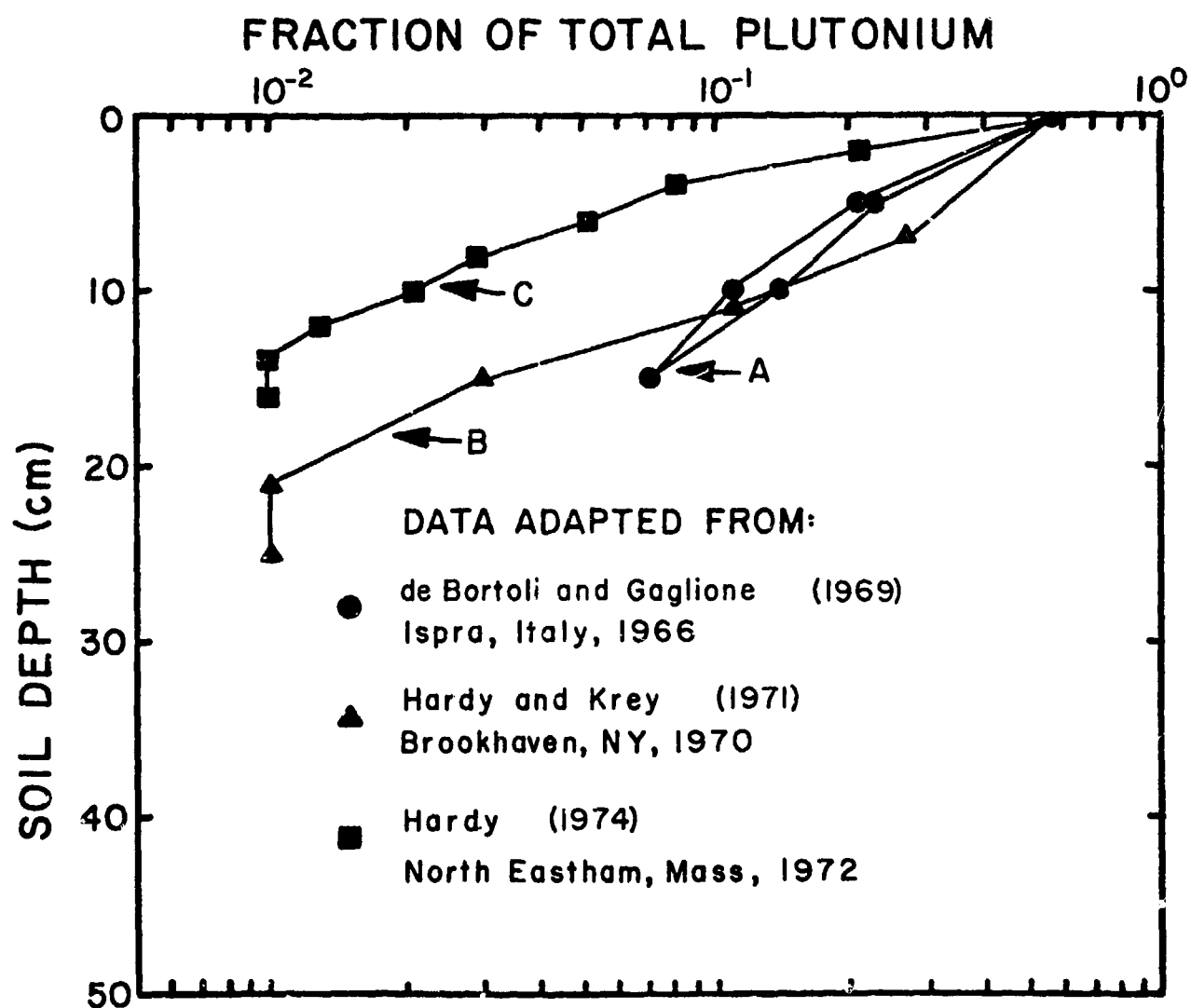


Fig. 1. Global Fallout Distribution in Sandy Soils.

To date no information has been found on ^{241}Am distribution in soils from global fallout. However, interest in the distribution of ^{241}Am in soils should be forthcoming, since the importance of ^{241}Am related to plant uptake and biological hazards has been recognized (Fowler and Essington, 1974).

Local Fallout

The Trinity event was the first atmospheric nuclear explosion. Plutonium distributions in soils near the Trinity Site, Alamogordo, NM, were reported for samples collected more than 20 years ago by Olafson *et al.* (1957). A 1972 study on this site was made by Hakonson and Johnson (1974) and plutonium distributions have been studied in more detail by Nyhan *et al.* (1976).

Olafson (1957) reported measurable downward plutonium movement occurring only where previous surface contamination was high and that the plutonium distribution followed the same trend as that found for fission products. Samples collected in 1948, three years after the event, showed the plutonium localized in the top 2 in. of soil. Hakonson and Johnson (1974) reported ^{238}Pu , ^{239}Pu distribution in soil profiles collected near Ground Zero (GZ) and at various distances along the fallout pathway to 56.4 km. Soil profile samples were obtained using a coring method resulting in 0- to 2.5-, 2.5- to 7.5-, and 7.5- to 30-cm increments. Positive total plutonium (^{238}Pu plus ^{239}Pu , ^{240}Pu) levels were found in the 7.5- to 30-cm increments in nearly all the profiles investigated out to 56.4 km. This suggests that some of the plutonium initially deposited 27 years ago had migrated to a depth of 7.5-30 cm in the soil. Except for the GZ profile, the plutonium concentrations in the 0- to 2.5-cm increments generally increased toward the outer end of the sampling transect. This observation is consistent with the fallout zone mapped by Olafson (1957), where the highest plutonium concentrations in soils, vegetation, and small mammals outside the GZ area occurred about 45 km from GZ. The vertical distribution of plutonium was relatively uniform in most of the core samples between GZ and the 24.1-km station. More distant profiles reflected a preponderance of plutonium in the surface 2.5 cm similar to the original observations of Olafson (1957) and Olafson and Larsen (1961). Hakonson and Johnson (1974) suggested that many factors could have accounted for Olafson's observations including differences with distance from GZ in the chemical and physical form of the plutonium and differences in the chemical, physical, and biological make-up of the environment.

Nyhan (1976) carried out a soil profile sampling program at Trinity in 1974 and 1975 to determine the maximum depth of ^{239}Pu , ^{240}Pu penetration and the variability of ^{239}Pu , ^{240}Pu penetration with location. Four sites were chosen: a control site 4.8 km south of GZ and three sites

1.6, 16, and 44 km northeast of GZ within the fallout pathway. Plutonium-239,240 at the control site, which represented global fallout, was too low to be detected below the 2.5- to 5.0-cm increment. On the other hand, $^{239,240}\text{Pu}$ was found to a depth of about 30 cm at the 44-km station. Figure 2 reproduces four of the profiles reported by Nyhan (1976) and is representative of the high degree of variability in $^{239,240}\text{Pu}$ distributions with depth. There was no consistent distribution pattern that could be related to obvious physical, chemical, or biological factors. The maximum depth of penetration, however, appeared to be related to the average maximum rain water penetration into the soil.

Nyhan (1976) observed that samples collected at greater distances from GZ reflected increased amounts of $^{239,240}\text{Pu}$ associated with $<100\text{ }\mu\text{m}$ -diam. particles. Various factors were noted as possible reasons for the increased $^{239,240}\text{Pu}$ migration at the 44-km site. This site receives 20% more rainfall than the other sites. Soils in this area contain gypsum, which could enhance the displacement of $^{239,240}\text{Pu}$ from soil binding sites due to the high concentration of soluble calcium ions and the enhanced water percolation due to flocculation of soil clays by the calcium ions. Another consideration was soil disturbance by the digging activities of badgers and other ground dwelling small mammals.

Atmospheric nuclear explosions also took place at NTS. Unfortunately, there is no record of measured transuranic nuclide distribution in soil profiles for any of those atmospheric nuclear explosions.

Two areas where atmospheric nuclear weapons testing has been conducted, and for which the vertical distribution of plutonium and americium has been measured, are the Enewetak and Bikini Atolls. Lynch and Gudiksen (1973) measured ^{239}Pu ($^{239,240}\text{Pu}$) and ^{241}Am in the coral soils and sands of Enewetak Atoll. Soil profile samples were collected to total depths of 35 to 185 cm. Concentrations of ^{239}Pu were reported in all profiles; ^{241}Am analyses were performed on many of the samples, but ^{241}Am data were not interpreted by the authors. Profile samples showed a wide range of activity distributions as a function of depth on different parts of the atoll, as shown in Fig. 3. Although meaningful generalizations regarding the ^{239}Pu distribution could not be made, Nervik (1973) commented on several of the groups of profiles observed. On the southern islands of the atoll, activity levels were usually low throughout the full range of depths sampled; some sampling locations showed ^{239}Pu concentrations decreasing somewhat from the surface through the first 10 or 20 cm of soil (Fig. 3, note trace A). Soil profiles inland on the islands subjected to fallout but not to construction or other GZ earth moving activities, showed a rapid and fairly steady decrease of ^{239}Pu levels from the surface to total depth, as shown in Fig. 3, trace B. In contrast, ^{239}Pu profiles from beaches and other exposed areas showed uniform or slowly decreasing ^{239}Pu activity levels from the surface to total depth (Fig. 3, trace C). Occasionally, distribution patterns showed accumulation of ^{239}Pu at

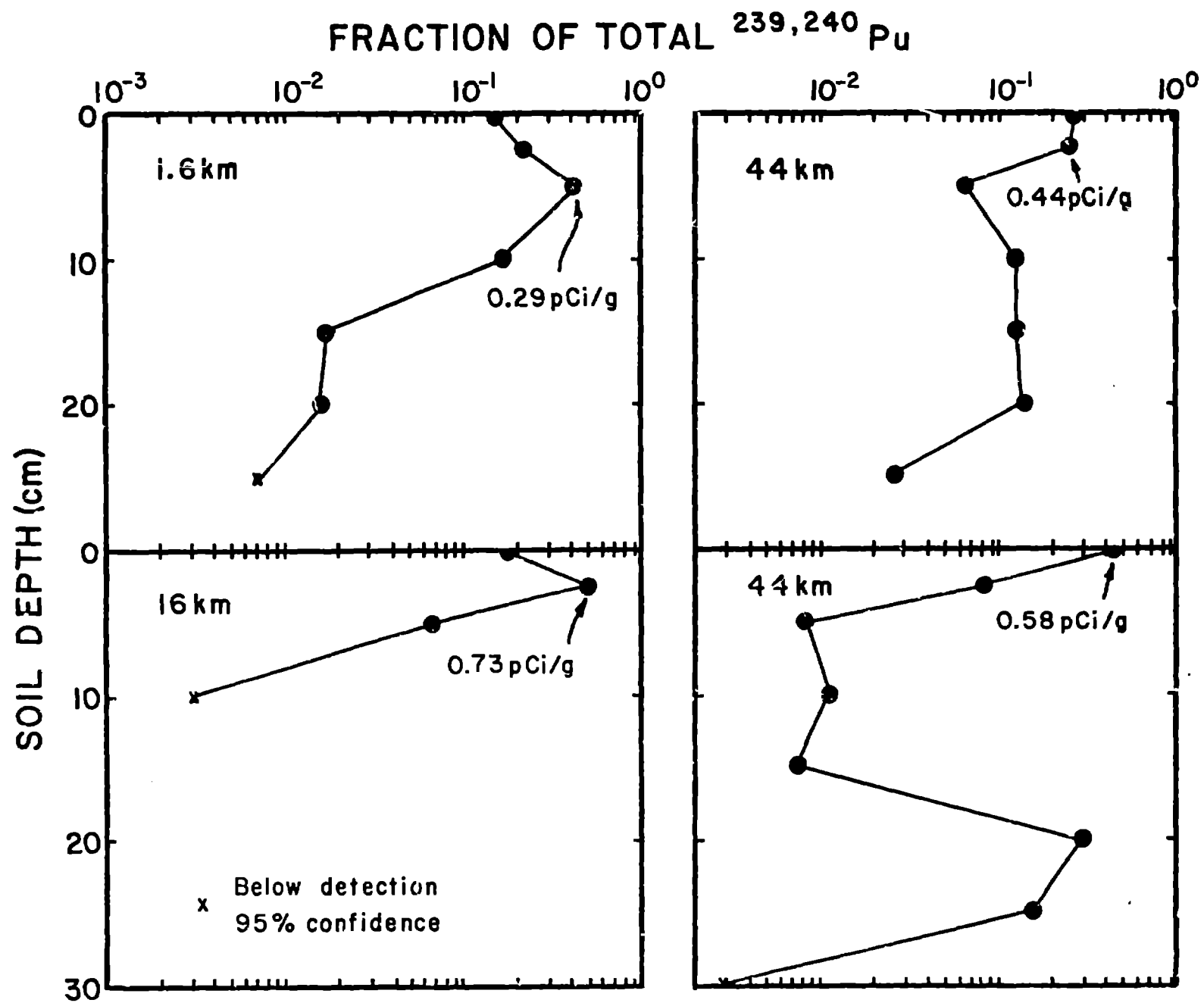


Fig. 2. $^{239,240}\text{Pu}$ Distribution in Trinity Site Soils
(Adapted from Nyhan *et al.*, 1976).

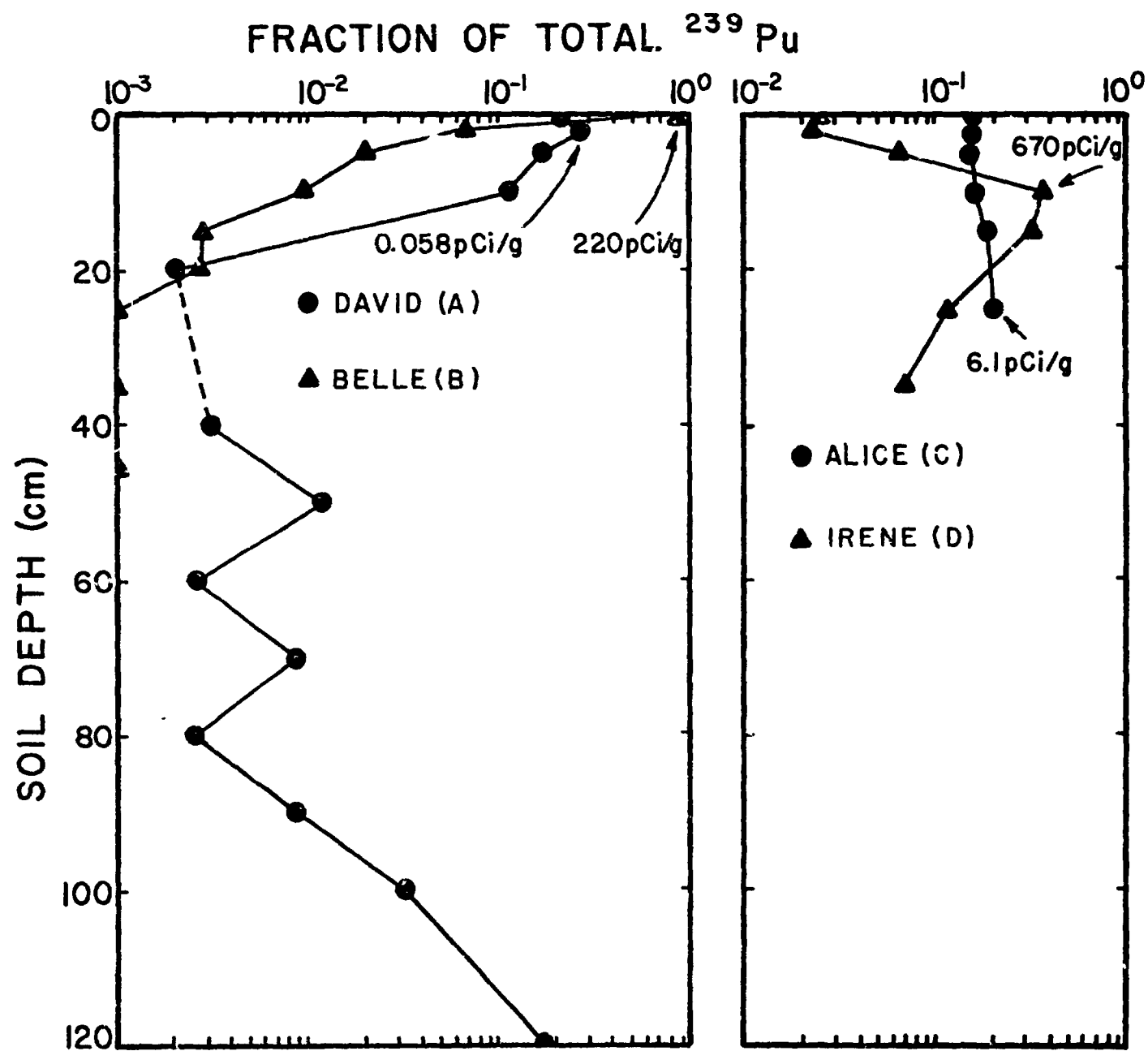


Fig 3. Distribution of ^{239}Pu in Enewetak Atoll Soils
(Adapted from Nervik, 1973).

some depth (Fig. 3, trace D); these profiles generally correlated with areas which had been subjected to testing, construction, or earth-moving activities. Several of the profiles indicated that ^{239}Pu may have migrated beyond the depth sampled, thus the total depth of penetration of ^{239}Pu remained unknown. A 1974 study by Noshkin *et al.*, (1976) of radionuclide concentrations in the ground water reservoir of one of the islands revealed that $^{239,240}\text{Pu}$ had migrated to a depth of 80 m below the island surface (Fig. 4). Plutonium-239,240 was detected at all intermediate depths sampled, suggesting that a portion of the $^{239,240}\text{Pu}$ was very mobile throughout the water-saturated coral-sand environments. As was shown in the soil profiles presented by Lynch and Gudiksen (1973), Noshkin (1976) also observed that the vertical distributions of $^{239,240}\text{Pu}$ at any one test well were unrelated to the distributions found on other islands or at different test wells on the same island.

Nevissi *et al.* (1976) discussed the distribution of $^{239,240}\text{Pu}$ in soils of Bikini Atoll. Plutonium-239,240 was detected as deep as 100 cm in soil profile samples with the distribution shown in Fig. 5. Trace A is a profile characteristic of a disturbed soil, possibly due to earth movement, construction, or clean up efforts. Traces B and C are two additional plutonium profiles from Bikini Atoll. It was noted that all the profiles exhibited $^{239,240}\text{Pu}$ distributions similar to those of ^{90}Sr measured on the same samples.

According to Nevissi (1976) there are three possible transport mechanisms for plutonium in Bikini soils: solution transport, biological transport, and suspension transport. Based on the soil and biological conditions at Bikini, it was concluded that solution transport and biological transport were not the major methods by which plutonium migrated downward. The suspension and resuspension of plutonium and plutonium-bearing particles was the principal mode of plutonium transport in these soils. Nevissi (1976) noted the observation of Held *et al.* (1965) that the algal crust of the atoll soils tend to retain a greater portion of all radionuclides. Although it was not possible to predict the mechanism of plutonium retention by the organic crust, the properties of the algal layers and the large adsorptive surface area may be one explanation. It was concluded that the present levels of $^{239,240}\text{Pu}$ and their distribution at Bikini, are not likely to change significantly in the immediate future unless mechanical disruption of the soil occurs.

Safety Shot Sites

A program is in progress under the auspices of NAEG to determine the inventory, distribution, and biological fate of transuranics and other prevalent radionuclides at the Safety Shot Sites of NTS. To date, this program has generated data on soil plutonium, americium, and uranium distributions for areas where safety tests of nuclear weapons have occurred.

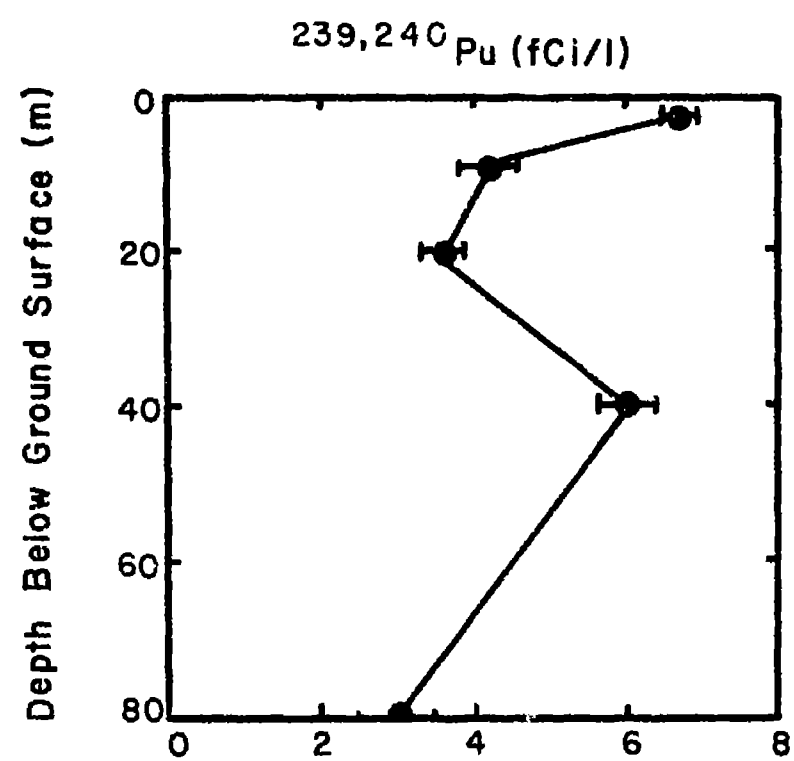


Fig. 4. Vertical Distribution of $^{239,240}\text{Pu}$ in a Test Well, Enewetak Atoll (Adapted from Noshkin et al., 1976).

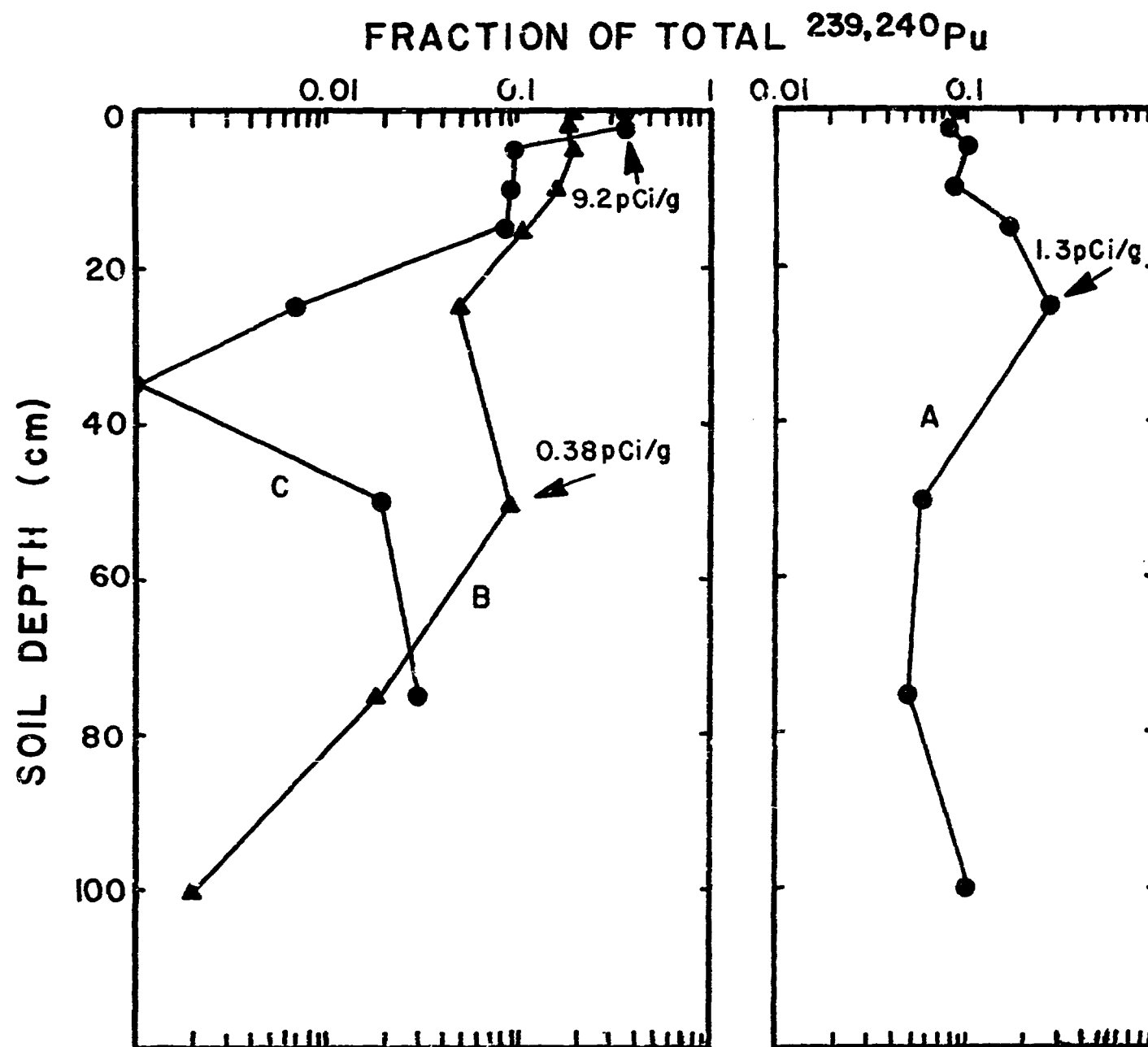


Fig. 5. Distribution of $^{239,240}\text{Pu}$ in Bikini Atoll Soils
(Adapted from Nevissi et al., 1976).

Romney *et al.* (1970) were the first to investigate the vertical distribution of ^{239}Pu ($^{239,240}\text{Pu}$) in the Safety Shot areas. Some ^{239}Pu was found to have migrated to 9- to 12-cm depths only 1.3 years after the event. Two profiles taken 10.8 years after the event showed similar distributions and indicated that deeper sampling would probably reveal additional ^{239}Pu penetration.

Between 1972 and 1973 a concerted effort was launched by NAEG to describe the inventory and distribution of uranium, plutonium, and americium at the Safety Shot Sites. This provided a more detailed evaluation of the vertical distribution of the nuclides. Essington *et al.* (1976) evaluated some 70 soil profiles from 7 test areas to determine the maximum depth of $^{239,240}\text{Pu}$ and ^{241}Am penetration and their distribution within the soil profile. As with the observations at Enewetak and Bikini, the $^{239,240}\text{Pu}$ distributions were extremely variable but could be categorized into three groups: normal, disturbed, and abnormal (altered by the physical and chemical conditions of the soil). Figure 6, trace A shows a normal or unaltered $^{239,240}\text{Pu}$ profile where the concentrations decreased with depth in a regular fashion described, perhaps, by a multiple exponential distribution. Figure 6, trace B shows a disturbed $^{239,240}\text{Pu}$ profile and differs from the Profile A in the large residual activity in the lower portions of the profile. This profile represents the $^{239,240}\text{Pu}$ distribution in a streambed area, where it is presumed that intense storm runoff has mixed the streambed materials and effectively distributed the $^{239,240}\text{Pu}$ throughout the depth sampled. Similar profiles were reported by Lynch and Gudiksen (1973) and Nevissi (1976) for highly sorted beach materials at Enewetak and Bikini Atolls. Other evidence of disturbance included: that of small mammal digs, post-event decontamination efforts, and the growth and reduction of blow sand mounds prevalent in the desert environment. Figure 6, trace C represents a rather widespread observation reflecting the effects of soil formation on $^{239,240}\text{Pu}$ distribution. The small increase in $^{239,240}\text{Pu}$ was correlated with the top of the "B" horizon, the zone of clay accumulation.

A very important aspect of plutonium migration into soils is the effect of time. Unfortunately, no experiments were designed nor were any measurements taken which would allow a detailed comparison of short and long term downward movement of plutonium. However, one of the profiles collected by Romney (1970) in 1958, 1.3 years after the safety shot event, was compared to a profile collected in 1973 by NAEG from a point 50 m away (Essington, 1976). The two profiles are shown in Fig. 7, normalized to fraction of total $^{239,240}\text{Pu}$ in the profile. The two profiles are similar, however the 1973 profile indicates a slightly deeper penetration of plutonium. The 1958 profile indicates that most of the plutonium found in the soil migrated shortly after deposition, and only a small amount may have moved to greater depths thereafter. In comparing these two profiles one must consider the sampling and analytical variability associated with each sample. Analytical variabilities (specifically radioactive counting statistics) for the 1973 samples are in the order of the symbol size; however, the variability for the 1958 samples was not given.

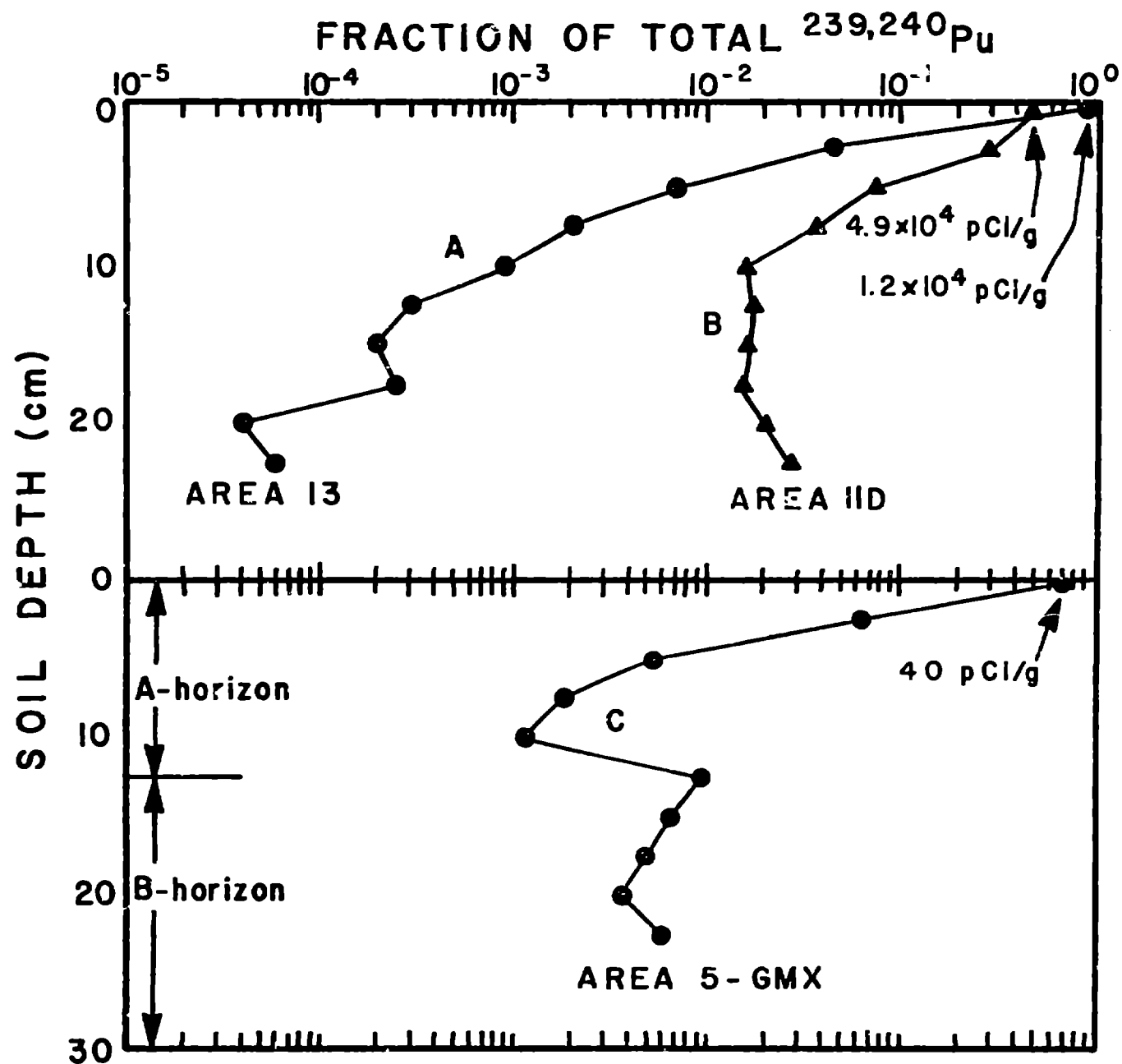


Fig. 6. Distribution of $^{239,240}\text{Pu}$ in Soils of the Nevada Test Site (Adapted from Essington et al., 1976).

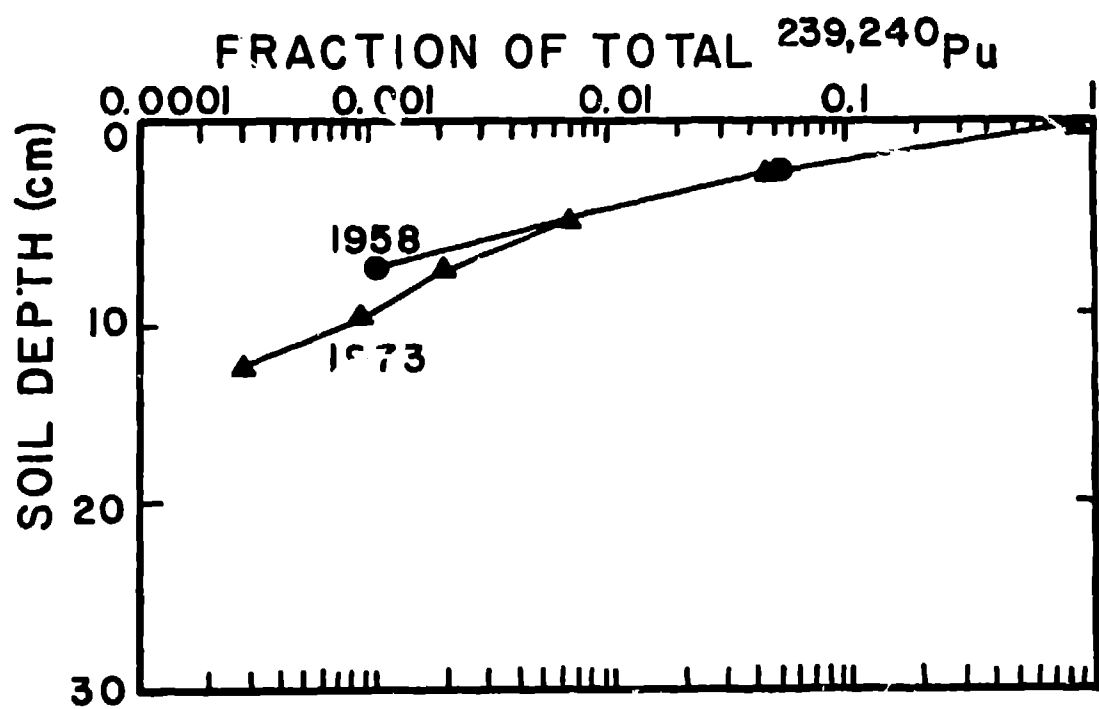


Fig. 7. Distribution of $^{239,240}\text{Pu}$ in Two Soil Profiles Collected in 1958 and 1973 at NTS (Adapted from Essington et al., 1976).

Very few measurements have been made of ^{241}Am movement in soils. Differential movement of americium and plutonium appears to have occurred in some of the soil profiles collected by NAEG. Essington (1976) presented a profile of $^{239,240}\text{Pu}/^{241}\text{Am}$ which indicated that small amounts of ^{241}Am appeared to be moving faster than $^{239,240}\text{Pu}$ at the Safety Shot sites. It should be pointed out, however, that not all of the profiles taken showed this pattern; ^{241}Am and $^{239,240}\text{Pu}$ appeared to move at the same rates in many of the profiles.

Accidental Releases of Plutonium

Fowler et al. (1968) reported results of soil sampling for plutonium at Palomares, Spain, after the accidental destruction of two nuclear weapons in that farming community. After considerable cleanup effort, a series of plots were located for the purpose of following the dispersion of the remaining low levels of plutonium from virgin land and from cultivated land. Figure 8 shows two such plutonium profiles which are the result of averaging nine fractional cores from each site. The effects of plowing the soil can be seen in the highly variable plutonium profile from the plowed field. A coring method was used to collect the soil profile samples from the two Palomares plots. In reviewing the data from this experiment it was noted that the distribution of plutonium predicted from the analytical results might be influenced by the cross-contaminating effect of the coring tool. Small particles of plutonium or plutonium oxide may have been dragged from the more radioactive surface soil into deeper profile samples, giving the appearance of a constant level of plutonium with depth.

The other major accidental release of plutonium for which soil profiles were investigated occurred at the Rocky Flats, CO, Nuclear Fuels Re-Processing Plant. A series of fires occurred over the years possibly releasing small amounts of plutonium to the environment, but probably the largest concentrated release occurred when drums filled with plutonium-laden oil and cuttings began leaking. To determine the distribution and amount of ^{239}Pu around the environs of the Rocky Flats plant, Krey and Hardy (1970) began a sampling program which included evaluation of the vertical distribution of ^{239}Pu ($^{239,240}\text{Pu}$) in the local soils. A number of soil profiles were taken in 1970, two of which are shown in Fig. 9. Plutonium-239 was found to a depth of 13 cm although the distribution was widely variable, presumably due to differences in soil chemical properties. In some cases as much as 60% of the ^{239}Pu was below 5 cm.

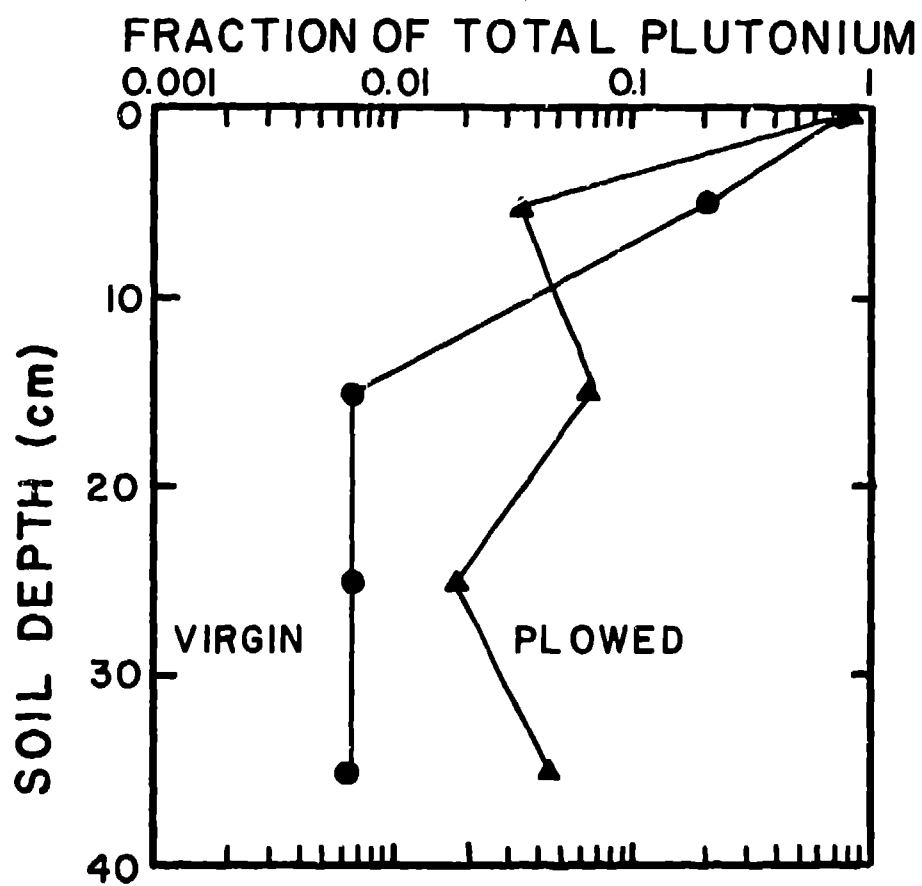


Fig. 8. Distribution of Plutonium in Palomares Soil
(Adapted from Fowler *et al.*, 1968).

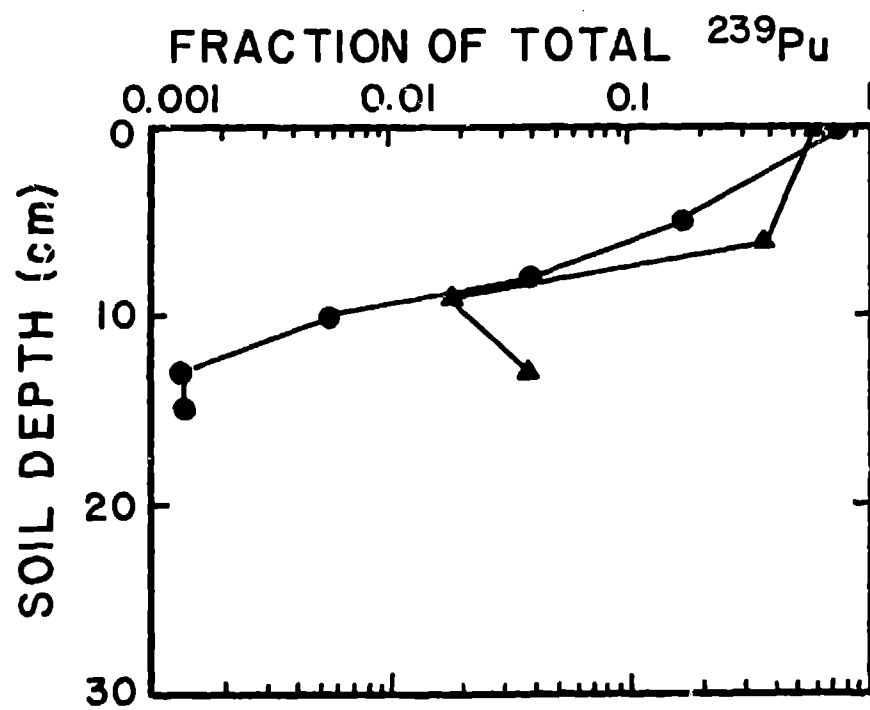


Fig. 9. Distribution of ^{239}Pu in Rocky Flats, Colorado Soil (Adapted from Krey and Hardy, 1970).

Special Studies

There have been a number of special studies dealing with migration of transuranics in soils, particularly with respect to laboratory investigations and radioactive waste disposal problems.

Routson *et al.* (1975) reported on a study to evaluate soils from eastern Washington and South Carolina in their capacity to sorb ^{237}Np and ^{241}Am . In this study the effects of Ca^{++} - and Na^{+} -ion concentrations on the radionuclide distribution coefficient, K_d , were determined. Distribution coefficient values for ^{237}Np decreased in all cases of increased Ca^{++} - and Na^{+} -ion concentrations, as shown in Fig. 10. Values for ^{241}Am also decreased with increasing Ca^{++} - and Na^{+} -ion levels in the South Carolina soil. However ^{241}Am - K_d values from the Washington soil were greater than 1200 and were not influenced by different concentrations of either Ca^{++} or Na^{+} .

The distribution coefficient can be related to ion migration rate in soil, making the proper assumptions that equilibrium is attained during leaching and that the species measured in the laboratory determination of K_d are the same as those in the leaching system and that the system is saturated. For example:

$$v_r = v_w \theta / [(1-\theta) \rho K_d + \theta]$$

where v_r = velocity of the radionuclide in soil,
 v_w = velocity of the leaching water,
 θ = fractional porosity,
 ρ = grain density, and
 K_d = measured distribution coefficient.

Using the above relationship and data of Fig. 10, ^{237}Np , as a soluble ion, can migrate much faster than ^{241}Am , particularly at low Ca^{++} - and Na^{+} -ion concentrations. Of course the specific physical and chemical characteristics of the soil will govern the actual degree of movement.

Glover *et al.* (1976) conducted equilibrium sorption measurements on 17 soils from the United States with plutonium and americium nitrates. This experiment was designed to discover relationships between plutonium and americium and certain physical and chemical characteristics of the soils. Among the factors investigated were cation exchange capacity, soluble carbonate, soluble salt content, organic carbon, pH, Eh, and particle size distribution. Distribution coefficient values for americium at a concentration of 10^{-10} M ranged from 82 to 10 000, and plutonium K_d values ranged from 35 to 14 000 for a concentration of 10^{-6} M. Regression analysis was used to determine correlations between chemical/physical parameters and the degree of radionuclide sorption. Cation exchange capacity and clay and sand contents were found to be the most important, which indicates that a conventional ion exchange process was responsible for the sorption of plutonium and

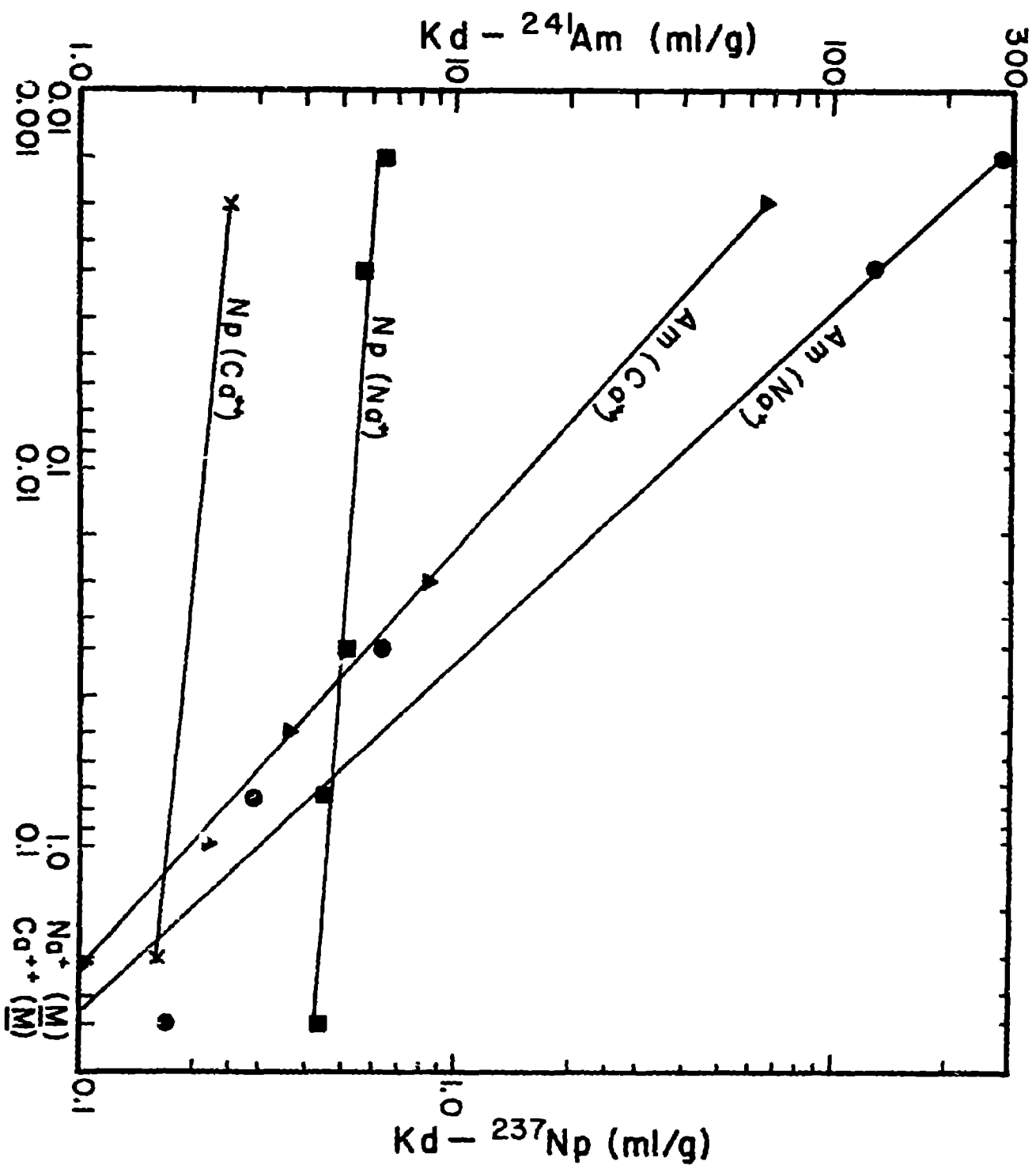


Fig. 10. ${}^{241}\text{Am}$ and ${}^{237}\text{Np}$ Distribution Coefficients (K_d) for South Carolina Soil (Adapted from Routsos et al., 1975).

americium. During the same study, elution characteristics of plutonium were determined in several of the soils for which K_d had been measured. Figure 11 shows the distribution of plutonium in three of those soils. As the K_d increased so did the retention of plutonium in the uppermost portions of the soil column.

Considerable amounts of ^{238}Pu are presently being used as heat sources for power generators particularly in space applications. Patterson et al. (1976) conducted an experiment to determine the fate of metallic ^{238}Pu under environmental conditions. Within environmental test chambers, a ^{238}Pu heat source specimen or fragments thereof, were placed on soils under simulated climatic conditions. The first "rain water" that percolated through the test soil contained a small amount of ^{238}Pu . This very rapid ^{238}Pu breakthrough indicated that some of the material that spilled from the fuel was very small particulate or colloidal. Auto-radiography of the percolated rain water revealed plutonium oxide particles of 0.05 to 0.8 μm diam. with a count median diameter of .095 μm . Nearly all of the plutonium oxide particles carried through the soil by water were less than .05 μm diam., except for the occasional presence of a particle as large as 4 μm . Patterson presented a number of soil profiles taken in the test chamber at the conclusion of the long-term leaching experiments. Using auto-radiography, it was observed that each plutonium oxide particle was associated with one microscopically visible soil particle. This indicated that trapping of plutonium oxide particles by soil was not a matter of filtration of the fine oxide particles, but that there was agglomeration of individual oxide and soil particles.

A recent, highly specialized, study on the distribution of actinides under waste disposal trenches at the Hanford Reservation in Washington was reported by Price and Ames (1976). This program was established to assess the future radiological impact of actinides in the ground underlying retired trenches and to develop methods for the long term control of contaminants. Samples were taken at 5 cm, 50 cm, 2 m, 4.5 m, and 9 m below the trench floor using a core sampler driven 0.5 m or 1 m for each profile increment. Both ^{239}Pu ($^{239}, ^{240}\text{Pu}$) and ^{241}Am were determined on the sediments from the core samples. One of the profiles is reproduced in Fig. 12. Examination of the core samples revealed both particulate and nonparticulate plutonium that may have been responsible for the observed distribution. The particulate fraction consisted of discrete plutonium particles, 2 to 25 μm diam., and was restricted to the top portions of the sediment columns. Price and Ames (1976) discussed in some detail hydrolysis and the possible soil-mineral-plutonium reactions which may have been responsible for sorption of plutonium in the lower profile fractions.

Similar coring studies by Holcomb et al. (1976) are in progress at Savannah River Plant.

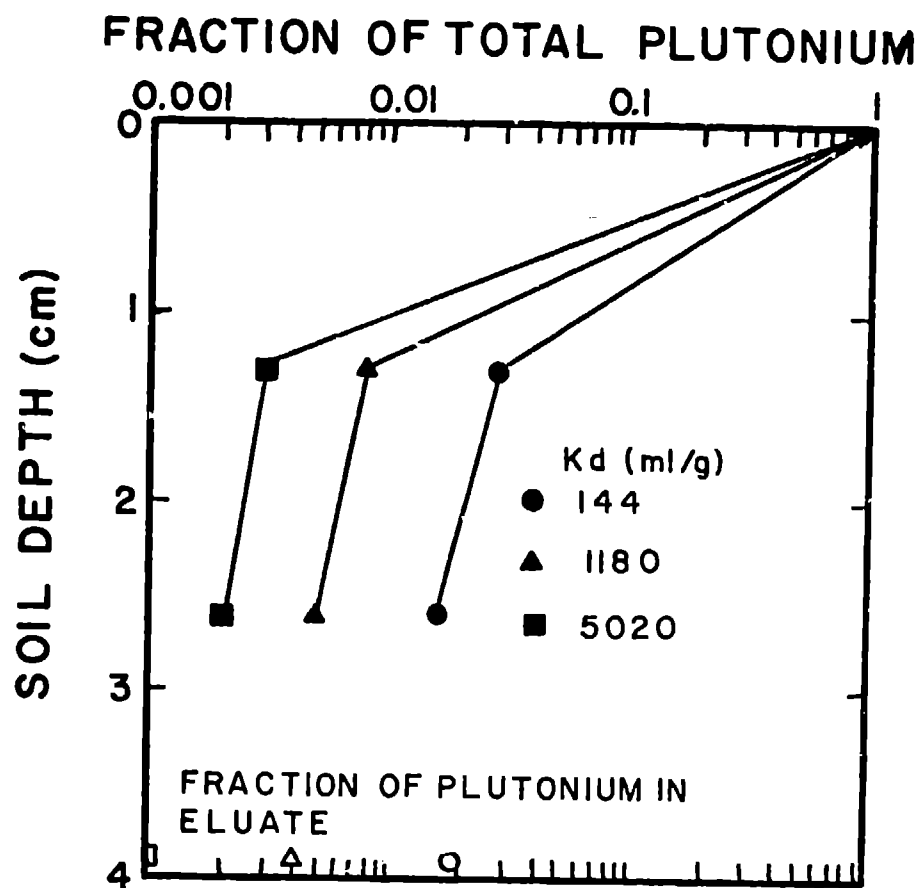


Fig. 11. Distribution of Plutonium in Soils Exhibiting Varying Plutonium Distribution Coefficients (K_d) (Adapted from Glover et al., 1976).

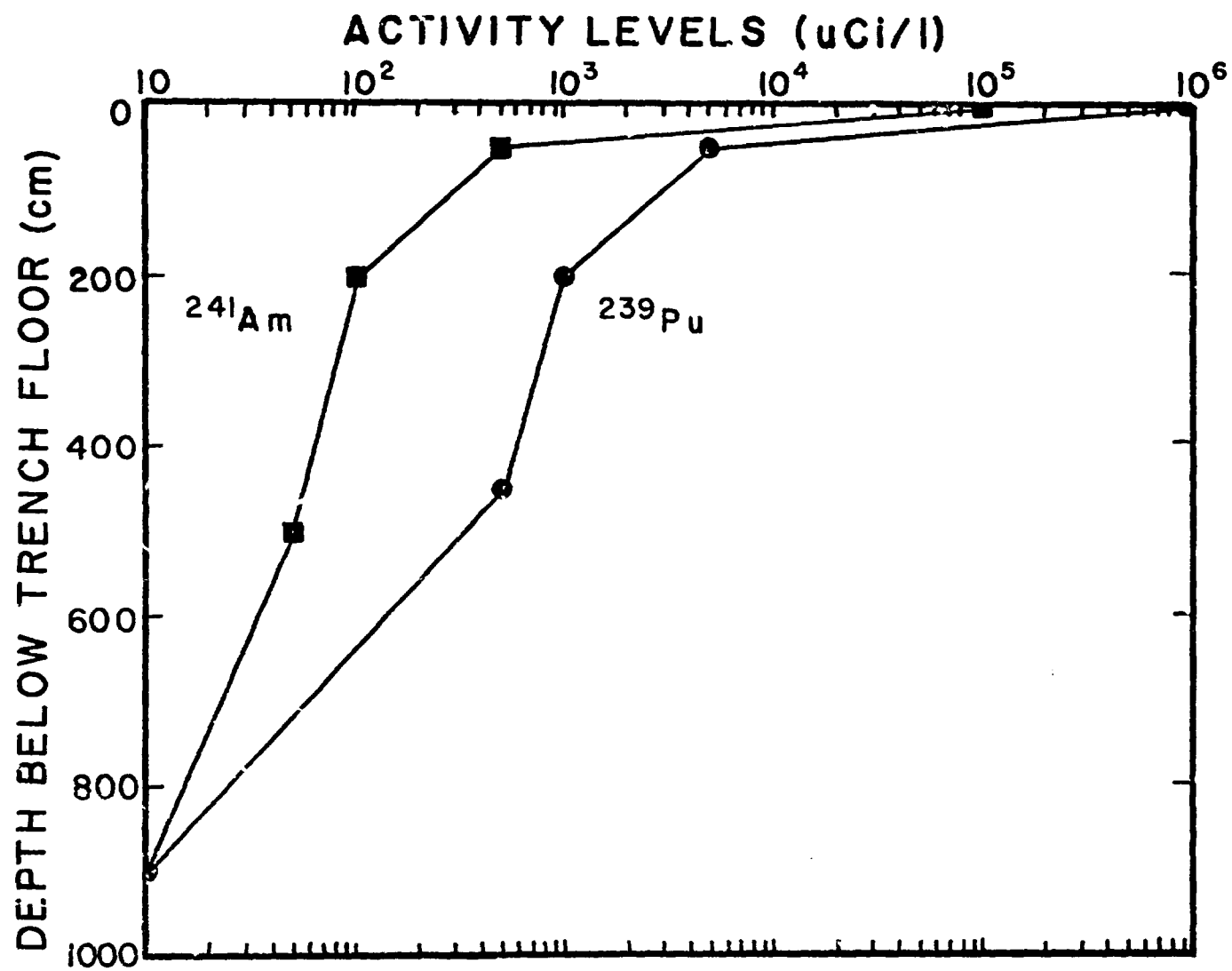


Fig. 12. Distribution of ²⁴¹Am and ²³⁹Pu Under Waste Disposal Trench at the Hanford Reservation, Hanford, Washington (Adapted from Price and Ames, 1976).

MOVEMENT OF URANIUM

Although uranium is not a transuranic, certain uranium isotopes in the nuclear fuel cycle have been identified as potentially hazardous and warrant discussion.

Till (1975) discussed the recycle aspects of ^{233}U bred from ^{232}Th in the High Temperature Gas-Cooled Reactors. The recycle ^{233}U can contain up to 1200 ppm ^{232}U , which was stated to be very radiotoxic, as having a relatively high specific activity, and as accounting for 82.1% of the total $^{232,233}\text{U}$ radioactivity. Table 3 lists the assumed activities of various uranium isotopes in 1 g of ^{233}U fuel. It can be seen that ^{232}U can account for greater than 50% of the total uranium activity present. Because of the potential environmental hazard of ^{232}U , an evaluation of uranium migration through soils is in order.

Natural uranium is distributed throughout minerals and sediments the world over. Hansen and Stout (1968) noted that uranium and thorium appeared to accumulate in soils as a result of rock weathering and soil formation. Maximum natural thorium concentrations tended to be in the "B" horizon, whereas the maximum uranium concentrations were most often in the topsoil. This suggests that thorium moves down the soil profile more rapidly than uranium. Generally, uranium and thorium concentrations are highest in the soil clay fractions; however, clays developed from carbonate rich soils show relatively low uranium and thorium concentrations. Hansen (1970), in a subsequent study, suggested that 600 000 years after initial deposition, most of the nuclides of the uranium and thorium families are in the finer textured soil fractions and that the uranium that is retained by the soil is more likely to be fixed in some immobile form.

Probably the only literature citation revealing uranium migration in environmental soils on a short-time basis was presented by Essington (1976). During the nuclear testing program discussed earlier in the section entitled "Safety Shot Sites," there was an explosive test of a ^{235}U enriched nuclear device. A soil profile taken from near the ground zero area (Fig. 13) showed the vertical distribution of $^{239,240}\text{Pu}$, ^{238}U , and ^{235}U . The ^{238}U levels reflected the natural concentrations in the mineral structure of the soil. However, when ^{235}U was distributed over the soil surface it did migrate into the soil profile as evidenced by the ^{235}U distribution. This added ^{235}U appeared to behave very much like $^{239,240}\text{Pu}$, also distributed at approximately the same time.

Table 3. Assumed Radioactivity of Uranium Isotopes
in 1 gram of ^{233}U Fuel

Isotope	Equilibrium recycle atom fraction	Equilibrium recycle activity (Ci)
^{232}U	.001	2.14×10^{-2}
^{233}U	.613	5.82×10^{-3}
^{234}U	.243	1.51×10^{-3}
^{235}U	.080	1.72×10^{-7}
^{236}U	.063	3.99×10^{-6}

Adapted from Till (1975).

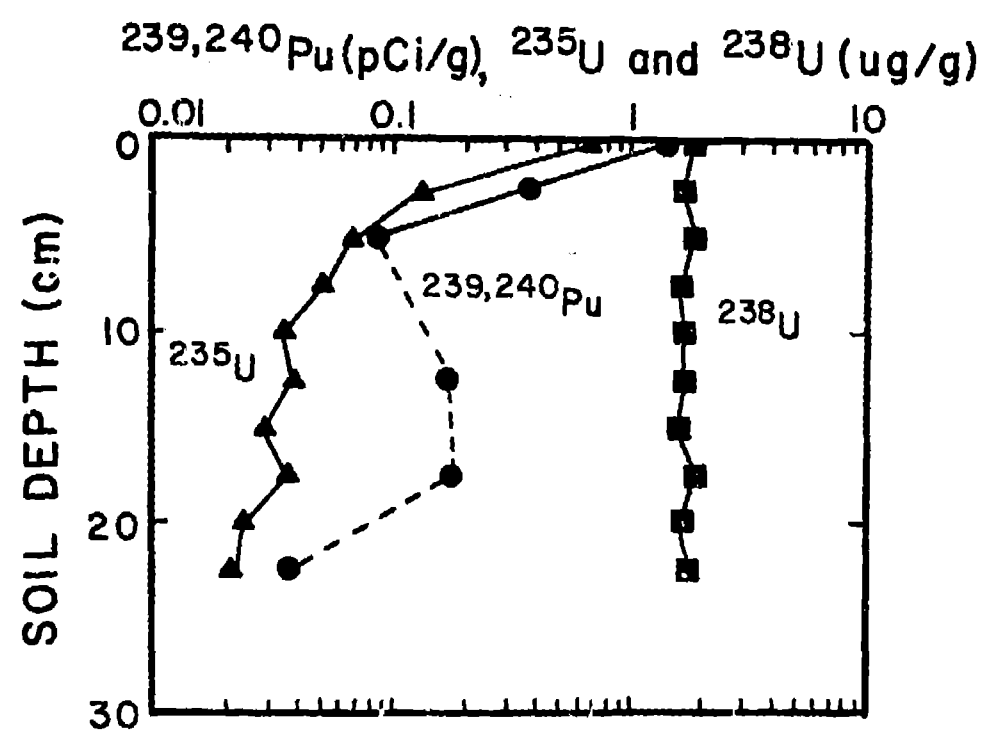


Fig. 13. Distribution of $^{239,240}\text{Pu}$, ^{235}U , and ^{238}U in NTS Area 11-A Soil (Adapted from Essington et al., 1976).

PREDICTIONS OF TRANSURANIC MOVEMENT IN SOILS

Attempts have been made to describe the movement of transuranics through soils on a mathematical basis. Lester et al. (1974) investigated the hypothetical band and impulse releases of transuranic parent-daughter chains through a soil column (that column which follows the ground water flow path) to a surface water body from an underground geologic nuclear waste disposal site. Numerical resolutions of analytical (mathematical) solutions revealed that differences in adsorption characteristics between soluble chain members, axial dispersion, and radioactive decay all act to reduce radionuclide discharge rates at the exit of the soil column. This method draws on a uniform, one-dimensional model and requires evaluation of the distribution coefficient, K_d , for each soil-radionuclide combination. Thus, this technique has limited application to predicting movement of transuranics in terrestrial soils where radionuclide K_d values can vary over wide ranges along the transport path. Other mathematical models to predict the distribution of radionuclides in soils have been presented by Cohen (1975) and Aleksakhin (1963).

A direct application of predicting transuranic distributions in a soil profile was presented by Jakubick (1976), who attempted to reconstruct plutonium concentrations in soils of south-west Germany. By correlating the few $^{239},^{240}\text{Pu}$ measurements available in Germany with the fallout data from Ispra, Italy, the probable history of plutonium fallout was reconstructed. From these data the plutonium input function in the vicinity of Heidelberg was derived. The plutonium distribution in a soil column from fallout in the United States was used in conjunction with the calculated cumulative fallout input function to predict the plutonium distribution and turnover rate in Heidelberg soils. Results of this study imply that the migration of fallout plutonium takes place completely, or to a great extent, in the form of discrete particles probably weakly aggregated to soil particles. The removal of fallout plutonium particles (analogous to plutonium oxide particles) from a given soil layer seemed to obey an exponential law, with a characteristic turnover time of 5-6 years for a 5 cm thick partially saturated soil layer.

DISCUSSION

This review has pointed out that there are many and diverse observations of transuranic distributions in soils; however, almost all of the observations are limited to plutonium. Some information on the distribution of americium has been produced, but generally only at nuclear weapons testing areas. Measurements on the distributions of the other

transuranics in environmental soils is lacking. Although the literature on transuranic chemistry in soils was not discussed, many of the papers reviewed either hypothesized, concluded, or in isolated cases measured the fact that plutonium had migrated into the soils and probably was in particulate form, either as plutonium oxide particles, hydrated plutonium oxide colloids, or plutonium particles aggregated to soil particles.

Even though the plutonium distributions in soil are quite variable, there has been an attempt at a rough categorization. It appears that irrespective of the source, plutonium profiles in soil can be categorized into three groups: normal, disturbed, and abnormal.

Normal profiles exhibit no obvious profile development and the plutonium distribution appears to reflect a complex exponential function. This type of profile is more likely to be associated with an undisturbed soil. Only a limited number of observed plutonium profiles can be categorized as normal.

The disturbed plutonium profiles result from activities associated with construction, burrowing animals, water and wind erosion, and redeposition after the initial deposition of plutonium. The plutonium distributions in these disturbed soils are quite varied. They might show a uniform distribution of plutonium with depth, that is, as deep as samples were collected, as was found on beaches and open areas of Enewetak and Bikini Atolls, or in a desert wash area where runoff from intermittent but severe rainstorms actively mix the top layers of material.

Abnormal plutonium distributions are generally unexplainable and a series of hypotheses have been used to suggest the possible mechanisms responsible for individual observed distributions. These possibilities range from faulty analyses of some fraction (or fractions) to unidentified mechanical disturbances, to the physical and chemical interactions of plutonium with the soil constituents. Understanding of the abnormal distributions may be a key to describing the physical and/or chemical interactions effective in the redistribution of plutonium or transuranic nuclides within soils. For instance, the accumulation of plutonium in specific soil zones which are related to soil forming factors, such as A and B horizons or carbonate lenses, has been recognized. The filtering action of specific soil horizons for small colloids containing plutonium or plutonium particles, and the potentially different chemistry among the zones, may be the long term governing factors for plutonium migration rates in soils. The authors are aware of no study currently in progress to address these problem areas.

Early in the history of plutonium distribution studies, it was thought that plutonium was so tightly bound to soil, that plutonium could not penetrate into the soil in the short time that has elapsed since its first introduction into the soil. This may be true for the migration of soluble ionic plutonium since it may be tightly bound by soil

particles. However, the migration of plutonium as soluble complexes, or in conjunction with soil particles also occurs to a sufficient degree, and some plutonium does migrate rapidly and to great depths, as evidenced by the ground water study at Enewetak.

Close examination of many of the published profiles indicates that the true front of plutonium migration was not observed due either to insufficient depth of sampling or inadequate detection capability. The implication is that a small amount of plutonium may have migrated to greater depths than those measured.

The few inferences that can be drawn from observations of the duration of migration suggest that for a short period of time after deposition onto soil there is a rapid penetration of plutonium into the soil. Indications are that a small amount of plutonium moves rapidly, and to some depth, into the soil as though the plutonium were complexed and relatively unaffected by the attractive forces of soil particles. For large concentrations of freshly deposited plutonium this effect may ultimately present a significant environmental hazard. Studies should be conducted to assess the magnitude of the possible mechanisms involved.

The turnover rate of fallout plutonium has been calculated to be 5-6 years for each 5 cm of depth giving rise to the exponential depth profiles in undisturbed soils. This observation appears to be somewhat independent of the total rainfall of the area.

Many of the early soil profiles were collected by coring techniques which may have introduced a cross-contamination variable, predicting higher-than-real concentrations of plutonium at depth. However, this problem was recognized and much of the later work used techniques (such as the "trench method") which provided some control over that variable. One cannot argue at this point with definitive data that the coring method is faulty, only that the variable does exist and remains unanswered.

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